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Title: IMS Rapid Response FY21 Summary Report for: Integrating Patterned Probes with Four-Dimensional Scanning Transmission Electron Microscopy for Unrivaled Crystallographic Structure Determination in Nanomaterials

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IMS Rapid Response FY21 Summary Report for:

Integrating Patterned Probes with Four-Dimensional Scanning Transmission Electron Microscopy for Unrivaled Crystallographic Structure Determination in Nanomaterials

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Goals/Objectives/State-of-the-Art. The initial goal of our 4-dimensional scanning transmission electron microscopy (4D-STEM)-based project was to develop strain resolution two orders of magnitude better than what is now currently possible with electron-based scattering techniques, all while collecting scattering information from 7 different tilt axes at one time [multi-beam electron diffraction (MBED)[1]] through the development of a new electron probe-forming aperture with non-circular features (patterned probes[2]). We set out to accomplish this through a collaboration with Drs. Colin Ophus and Ben Savitsky at Lawrence Berkeley Laboratory (they are the world-leading experts in developing the complex computational codes required to perform orientation analysis and quantitative strain mapping on our 4D-STEM data sets. We are motivated to invest in this area as it will be the only technique sensitive enough to perform three-dimensional automated crystallographic orientation mapping (ACOM) and strain mapping for materials exposed to external stimulus (a focus of our larger efforts).

Connection to LDRD Funding Mission. ***Capability.*** We implemented patterned-probe 4D-STEM at LANL through this project. We submitted and were awarded 2 user proposals with the National Center for Electron Microscopy (resides in LBL's Molecular Foundry) regarding collaboration on custom electron-probe forming apertures. After we executed two Material Transfer Agreements between LANL and LBL, we obtained a total of 3 'bullseye' apertures. One was installed on the Titan TEM in the LANL Electron Microscopy Laboratory, and one was installed on the new TEM in TA-55 but broke so we just received a new replacement aperture which will be installed in FY22. We also submitted and were awarded 3 additional user proposals with LBL Molecular Foundry to obtain codes and analysis software for performing orientation mapping in the TEM, this was implemented remotely through 3 hands-on 'virtual' tutorials with Dr. Colin Ophus which were attended by ~5-10 LANL personnel. We did not get to the step of MBED aperture installation as those are still not available, and this will be a follow-on project after we publish our orientation mapping results. We published 3 papers from this project and have one paper which will be submitted in early FY22[3-6]. ***Mission Agility.*** Our team was awarded a new Mission Foundations Research award starting FY22 to continue to implement 4D-STEM at LANL for characterization of actinides, "*LDRD MFR 20220485 Phase 1: Coupling Multiple Patterned Electron Probes for Real Time Orientation, Lattice Parameter, and Strain Mapping at the Nanoscale*." ***People.*** We have also used the techniques developed in this IMS RR award in an existing DR led by Abby Hunter and Saryu Fensin which has enabled a new collaboration for our team, "*LDRD DR 20210036DR: Investigating How Material's Interfaces and Dislocations Affect Strength (iMIDAS)*." The techniques developed in this project also helped our project postdoc Dr. Alejandra Londoño-Calderon be recruited as a senior TEM analyst at Applied Materials where she was hired to implement the 4D-STEM techniques we developed in an industrial setting using probe-corrected TEMs. We also added 2 director's funded postdocs to our LBL MF proposals [Dr. Hi Vo (MST-8) and Dr. Xuejing Wang (MPA-CINT)] so that they can travel to collect experimental data for follow-on work.

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